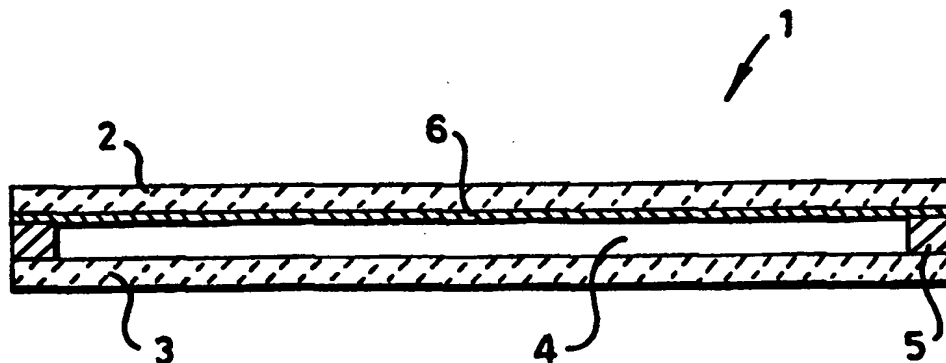




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<p>(21) International Application Number: PCT/GB98/00407</p> <p>(22) International Filing Date: 10 February 1998 (10.02.98)</p> <p>(30) Priority Data: 9702777.5 11 February 1997 (11.02.97) GB</p> <p>(71) Applicant (for all designated States except US): PILKINGTON PLC [GB/GB]; Prescott Road, St. Helens, Merseyside WA10 3TT (GB).</p> <p>(72) Inventors; and (75) Inventors/Applicants (for US only): STANLEY, Andrew, Thomas [GB/GB]; 30 Latham Avenue, Ormskirk, Lancashire L39 2EU (GB). RAMSEY, Philip, Michael [GB/GB]; Flat 3, Northfield, 12 Park Road West, Southport, Merseyside PR9 0JS (GB).</p> <p>(74) Agent: HALLIWELL, Anthony, Charles; Pilkington plc, Pilkington Technology Centre, Group Patents Dept., Hall Lane, Lathom, Ormskirk, Lancashire L40 5UF (GB).</p>	<p>(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, GW, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).</p> <p>Published With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</p>	

(54) Title: IMPROVEMENTS RELATING TO SOLAR CONTROL GLASS AND GLAZING



(57) Abstract

A multiple glazing unit comprising an outer pane and an inner pane, the outer pane being of tinted glass having a total iron content (calculated as ferric oxide) in the range 0.3 to 2 % by weight, and carrying an infra red reflecting metal oxide coating on its surface facing the interior of the unit. The outer pane may be of neutral tinted glass whereby the unit and the pane each have a total solar heat transmission of at least 5 % less than the visible light transmission and/or may comprise a tinted glass having a coating which comprises an iridescence suppressing layer or layers and a pyrolytic fluorine doped tin oxide layer having a thickness in the range 200 nm to 600 nm.

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IMPROVEMENTS RELATING TO SOLAR CONTROL GLASS AND GLAZING

The invention relates to glazing, and in particular to a high performance solar control multiple glazing unit and a novel coated neutral tinted glass for use in the construction of such a unit.

High performance solar control double glazing units which rely on complex silver coatings for their performance are known. One such unit, available on the market in the USA utilises a coating comprising two thin silver layers sandwiched between dielectric layers on surface two of the unit (the surfaces of a double glazing unit are commonly referred to as surfaces 1, 2, 3 and 4, being numbered from outside to in; thus surface two is the inward facing surface of the outer pane of the unit).

This unit has a high light transmission and relatively low heat transmission, with a neutral tint in both reflection and transmission, resulting from the use of the complex silver coating. Unfortunately, the complex silver coating is not suitable for deposition at atmospheric pressure and must be deposited under vacuum, for example, by magnetically enhanced sputtering, carried out after production of the glass. It would be desirable to have a high performance neutral coloured unit with a high light transmission that could be constructed of glass produced on line without the need for a subsequent, off-line, coating step.

We have now found that a comparable performance may be achieved by using, as the outer pane of the unit, a tinted glass pane having a composition including an iron content (calculated as Fe_2O_3), preferably in the range 0.3 to 1% and preferably with additional colouring additives to achieve a neutral colour, and an infra red reflecting metal oxide coating, suitable for on-line application, on one surface thereof.

Thus, according to the present invention, there is provided a multiple glazing unit comprising an outer pane and an inner pane, the outer pane being of neutral tinted glass having total iron content (calculated as ferric oxide) in the range 0.3 to 2%, preferably 0.3 to 1%, by weight, and carrying an infra red reflecting metal oxide coating on its surface facing the interior of the unit, whereby said unit has a total solar heat transmission at least 5%, preferably at least 10%, less than its visible light transmission.

The light transmission is measured using C.I.E. Illuminant C over the wavelength range 380 nm to 780 nm at 10 nm intervals; the total solar heat transmission (TSHT) is the total (i.e. directly transmitted heat plus absorbed and re-radiated heat) solar heat transmitted at Air Mass 2 (simulating rays from the sun incident at an angle of 30°) measured over the wavelength range 350 to 2100 nm at 50 nm intervals. All the transmissions referred to in the present specification and claims are calculated by applying the rectangular rule to the measured values.

UK patent specification GB 2 068 442A relates to a window assembly including means for mounting two panes of glazing material in a frame so as to be reversible as a body, so that either pane can be selectively positioned as the outer pane. In accordance with the specification windows can be positioned in preferred summer and winter orientations. In certain embodiments, described with reference to Table 8 of the specification, one of the panes of a double glazing panel is grey or bronze tinted glass with a fluorine doped tin oxide coating on its internal surface and the second pane is of clear float glass. In the winter orientation, with the tinted pane inwards, these units provide a visible light transmission of 30.2% (grey glass) or 34.9% (bronze glass) and a total solar heat transmission of 64.3% (grey glass) or 64.5% (bronze glass); changing to the summer orientation, with the tinted pane outwards, does not affect the visible light transmission, but significantly reduces the total solar heat transmission to 34.5% (grey glass) or 35.4% (bronze glass). It will be noted that none of these units, whatever its orientation, provides a heat transmission lower than its visible light transmission. Such a result is, admittedly, provided by the second unit of Table 7 but this employs a green tinted glass and therefore does not meet the requirement for a neutral tinted product.

By neutral tint, we mean exhibiting a transmission colour having a^* and b^* values such that $\sqrt{a^{*2} + b^{*2}}$ is less than 10, and preferably less than 8. Even lower values are preferred, although values of less than 7 may be more difficult to achieve, especially when a low (less than 50% total solar energy transmission) is required. However, when such a low total solar energy is not critical, values of 5 or less may be fairly readily achieved using thin (e.g. up to about 4 mm) tinted glass.

Although UK patent specification GB 2 068 442A does not specify the compositions of the tinted glasses used, it is believed that the improved performances of the neutral tinted

glasses of the present invention result from higher concentration of iron in the range 0.3 to 1% by weight (calculated as Fe_2O_3) in the glass.

The tinted glasses used in the present invention generally have a base glass composition in the range:

68 - 75% SiO_2
0 - 5% Al_2O_3
10 - 18% Na_2O
0 - 5% K_2O
0 - 5% MgO
5 - 15% CaO
0 - 5% B_2O_3
0 - 5% BaO
0.1 - 0.3% SO_3
0 - 3% ZrO_2

with 0.3 to 2%, preferably 0.3 to 1.1% and especially 0.3% to 1% iron (calculated as ferric oxide) and additional colourants to neutralise the colour of the glass. The glass may also contain other additives, for example, refining aids, which would normally be present in an amount of up to 2%. Preferably, the additional colourants used are selenium (typically in an amount of from 0.5 to 20 ppm, preferably 0.5 - 10 ppm), and cobalt (typically in an amount, calculated as Co_3O_4 , of from 8 to 100 ppm, preferably 8 - 40 ppm), although other colourants, for example nickel oxide and/or titanium oxide, may also be used if desired. Cerium oxide may be used to further reduce the ultraviolet transmission. The glass compositions specified in the present specification and claims are all defined in percentages or parts by weight, based on the total weight of the composition (including both base glass and colourants).

The ratio of ferrous iron to total iron will depend on the redox conditions present during the glass making process, and will normally be in the range 0.18 to 0.40, especially 0.18 to 0.35, (and preferably 0.20 to 0.30) with the higher ratios providing the best performance, i.e. greatest difference between visible light transmission and solar heat transmission. Thus the ratio will usually be greater than 0.20 and preferably greater than 0.25.

The total iron content is specified in weight percent ferric oxide (Fe_2O_3) assuming all the iron is present as ferric oxide. References to the ratio of ferrous iron to total iron in the present specification and claims are references to ratios determined optically and given by the following formula, in which $T_{1000\text{nm}}$ is the percentage transmission of radiation of wavelength 1000 nm through a sample of glass L mm thick, and Fe_2O_3 is the total iron content of the glass (calculated as ferric oxide) as the percentage by weight.

$$\text{Ratio ferrous iron/total iron} = \frac{\left[\log_{10} \left(\frac{100}{T_{1000\text{nm}}} \right) - 0.036 \right] 1.152}{L \times \text{Fe}_2\text{O}_3}$$

The resultant value represents the proportion of the iron content which is present in the ferrous (as opposed to ferric) state.

Units with a light transmission of at least 50%, especially at least 55%, are preferred, and to achieve these high light transmissions the tinted glass used will normally have an iron content of less than about 1.3%, especially less than about 1%, for 3 mm glass, reducing with increasing pane thickness to less than about 0.9%, especially less than about 0.7%, for 6mm glass (and reducing further as the tinted pane thickness increases). Moreover, it is preferred that the multiple glazing units of the present invention have a total solar heat transmission of not more than 50%.

The infra-red reflecting metal oxide coating on the tinted pane will normally be a doped metal oxide, for example a doped tin oxide or doped indium oxide coating, typically of such thickness and properties that the coated tinted glass exhibits an emissivity of less than 0.2. Not only are such metal oxide coatings generally more durable than the prior art silver coatings discussed above, but they are more suitable for production by an on-line coating process applied to the ribbon of glass as it is produced, so avoiding the need for a separate, off-line, coating step.

It is especially preferred to use, for the infra red reflecting metal oxide coating, a fluorine doped tin oxide layer having a thickness in the range 200 nm to 600 nm, preferably 300 nm to 500 nm, with an iridescence suppressing underlayer or underlayer(s). Such coating layers are applied commercially by pyrolytic coating processes applied on-line to a ribbon of glass as it is produced and have the advantage of durability referred to above. The

iridescence suppressing underlayer(s) may be as described in patents of Professor Gordon, for example GB 2 031 756B.

The preferred infra red reflecting metal oxide coatings used in the present invention are usually used on clear glass to retain heat (as, for example, in the use of the commercially available products ENERGY ADVANTAGE™ glass and PILKINGTON K GLASS™) but the present invention employs them to reject heat. The tinted glass to which the coatings are applied in accordance with the practice of the present invention is preferably a neutral tinted glass, but the use of the particularly preferred infra red reflecting metal oxide coatings referred to above on more highly coloured tints (i.e. tints not conforming to the definition of "neutral" set out above) is also new and especially advantageous when these coated tinted glasses are employed in multiple glazing units with the tinted glass used as the outer pane of a multiple glazing unit.

Thus, according to a further aspect of the present invention, there is provided a multiple glazing unit comprising an outer pane and an inner pane, the outer pane being of tinted glass carrying a coating on its surface facing the interior of the unit wherein said coating comprises an iridescence suppressing layer or layers and a pyrolytic fluorine doped tin oxide layer having a thickness in the range 200 nm to 600 nm.

The unit and the tinted glass outer pane used in it may each desirably have the features discussed above and/or claimed in any of the dependent claims.

To achieve the desired performance, the multiple glazing units of the present invention may be constructed using, for the outer pane, a pane of neutral tinted glass having an iron content (calculated as Fe_2O_3) in the range 0.3 to 2%, preferably 0.3 to 1%, by weight carrying an infra-red reflecting metal oxide coating on one surface thereof and having a total solar heat transmission at least 5% less than its light transmission. Such neutral tinted coated glass is novel and constitutes a further part of the present invention.

Preferably, the neutral tinted coated glass has a total solar heat transmission at least 10% less than its light transmission.

Moreover, because of the durability of the coatings, panes of the coated neutral tinted glass of the present invention may, if desired, be glazed monolithically, without the need for incorporation in a multiple glazing unit.

When the panes of coated neutral tinted glass used in the present invention have a thickness in the range 3 to 5 mm, they will normally have an iron oxide content in the range

0.5 to 0.9%, preferably 0.5 to 0.8% by weight; thicker panes, i.e. having a thickness of at least 5 mm, will normally have an iron oxide content in the range 0.4 to 0.8%, preferably 0.4 to 0.6% by weight.

The invention is illustrated but not limited by the following description with reference to the accompanying diagrammatic drawing of a section through a double glazing unit in accordance with the invention.

Referring to the drawing, a double glazing unit 1 comprises opposed parallel spaced panes 2 and 3, separated at their periphery by, and with gas space 4 between them sealed by, spacing and sealing means generally designated 5.

Pane 2, which is to be the outer pane when the unit is glazed, comprises a neutral tinted glass having a total iron content in the range 0.3 to 2% by weight and carrying an infra red reflecting metal oxide coating 6 on its surface towards the gas space 4, whereby the unit has a total solar heat transmission at least 5% less than its visible light transmission. Inner pane 3 is of clear float glass.

In a first series of Examples, pane 2 is composed of neutral tinted glass comprising the following base glass composition:

SiO ₂	72.5%
Al ₂ O ₃	1.0%
Na ₂ O	12.9%
K ₂ O	0.6%
CaO	8.3%
MgO	3.9%
SO ₃	0.2%

with the following colourants:

Total iron (calculated as Fe ₂ O ₃)	0.50%
Ratio ferrous iron (calculated as Fe ₂ O ₃) to total iron (calculated as Fe ₂ O ₃)	0.20
Cobalt (calculated as Co ₃ O ₄)	15 ppm
Selenium (calculated as selenium metal)	2.5 ppm

all parts and percentages being by weight, based on the total (base glass + colourants) weight of the glass.

The first pane in the series (Example 1) is 3 mm thick; the second pane (Example 2) is 6 mm thick. Each tinted pane carries a pyrolytic infra red reflecting fluorine doped tin oxide coating of approximately 350 nm thickness providing a normal emissivity of 0.16 applied over a colour suppressing underlayer of the kind described in UK patent GB 2 031 756B.

The basic optical properties of the panes are set out in Table 1a. Table 1b shows, for each of the tinted panes of Examples 1 and 2, the performance of a double glazing unit as illustrated in the drawing in which the inner pane is a pane of clear float glass of the same thickness as the outer pane, with the gap between the panes 12mm wide and filled with air.

Corresponding values for similar double glazing units, but with the coating on the air space side of the inner clear float pane and the outer tinted pane uncoated are shown in brackets in Table 1b by way of comparison.

Table 1a

	Example 1	Example 2
Visible light transmission	72%	61%
Total solar heat transmission	59%	50%
Transmission colour a*	-3.2	-5.7
b*	+2.9	+3.4
$\sqrt{a^{*2} + b^{*2}}$	4.3	6.6

Table 1b

	Example 1 (double glazing unit)	Example 2 (double glazing unit)
Visible light transmission	66% (66%)	55% (55%)
Total solar heat transmission	51% (55%)	42% (46%)
Transmission colour a*	-3.9	-6.5
b*	+2.7	+3.2
$\sqrt{a^{*2} + b^{*2}}$	4.7	7.2

The data in Tables 1a and 1b shows the high performances achieved, even with a neutral coloured glass, when using an outer tinted pane with a total iron content of 0.5% and

an infra red reflecting coating, the heat transmission being, in each case, substantially lower than the light transmission. It is apparent, from the figures in brackets, that the performance, in particular in respect of heat transmission, of the double glazing unit is substantially improved by using the low emissivity coating on the outer tinted pane, rather than on the clear glass inner pane.

In a further series of Examples, pane 2 is composed of a neutral tinted glass having the same base glass composition as in Examples 1 and 2, but with the following colourants:

Total iron (calculated as Fe_2O_3)	0.45%
Ratio ferrous iron (calculated as Fe_2O_3) to total iron (calculated as Fe_2O_3)	0.26
Cobalt (calculated as Co_3O_4)	10 ppm
Selenium (calculated as selenium metal)	2.5 ppm

wherein, as before, all parts and percentages are by weight, based on the total (base glass + colourants) weight of the glass.

The first pane in this series (Example 3) is 3 mm thick; the second pane (Example 4) is 6 mm thick. Each tinted pane carries a pyrolytic infra red reflecting fluorine doped tin oxide coating of approximately 350 nm thickness providing a normal emissivity of 0.16 applied over a colour suppressing underlayer of the kind described in UK patent GB 2 031 756.

Tables 2a and 2b set out the optical properties of the panes (Table 2a) and double glazing units incorporating the panes (Table 2b). In each case, the double glazing units are as shown in the drawing, with the inner pane being a pane of clear float glass of the same thickness as the outer pane, and the gap between the panes 12mm wide and filled with air. The figures shown in brackets in Table 2b are the corresponding values for modified units using the same outer panes but in uncoated form, with the infra red reflecting coatings on the air space sides of the inner clear float panes.

Table 2a

	Example 3	Example 4
Visible light transmission	72%	61%
Total solar heat transmission	58%	48%
Transmission colour a*	-3.2	-5.7
b*	+2.9	+3.4
$\sqrt{a^{*2} + b^{*2}}$	4.3	6.6

Table 2b

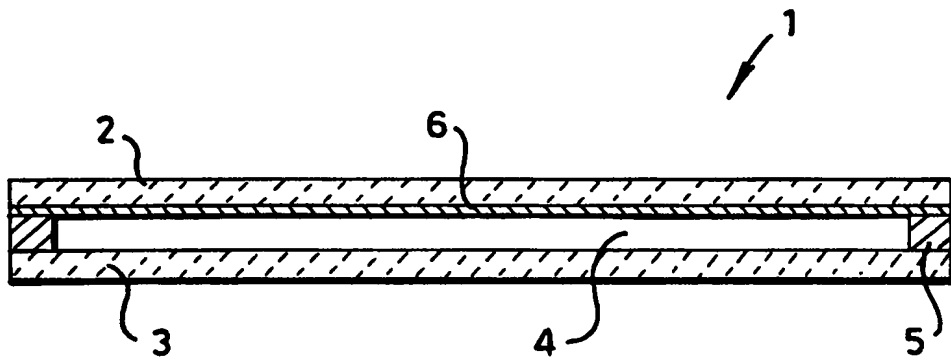
	Example 3 (double glazing unit)	Example 4 (double glazing unit)
Visible light transmission	66% (66%)	55% (55%)
Total solar heat transmission	50% (53%)	40% (44%)
Transmission colour a*	-3.9	-6.4
b*	+2.7	+3.2
$\sqrt{a^{*2} + b^{*2}}$	4.7	7.2

By comparing Example 1 with Example 3, and/or Example 2 with Example 4, it will be seen that the performance of both the coated tinted glass, and double glazing units comprising the glass, in selectively admitting light rather than heat, has been improved still further within the scope of the invention by modification of the glass composition. Thus the visible light transmission values (and the colours) have remained unchanged, while the heat transmission values have been reduced by 1% for the 3 mm glass cases (Example 3) and by 2% for the 6 mm glass cases (Example 4).

Claims

1. A multiple glazing unit comprising an outer pane and an inner pane, the outer pane being of neutral tinted glass having total iron content (calculated as ferric oxide) in the range 0.3 to 2% by weight, preferably 0.3 to 1%, by weight, and carrying an infra red reflecting metal oxide coating on its surface facing the interior of the unit, whereby said unit has a total solar heat transmission at least 5% less than its visible light transmission.
2. A multiple glazing unit according to claim 1 having a solar heat transmission at least 10% less than its visible light transmission.
3. A multiple glazing unit according to claim 1 or claim 2 having a visible light transmission of at least 50%.
4. A multiple glazing unit according to claim 3 having a visible light transmission of at least 55%.
5. A multiple glazing unit as claimed in any of the preceding claims having a total solar heat transmission of not more than 50%.
6. A multiple glazing unit as claimed in any of the preceding claims wherein the coated outer pane has a total solar heat transmission at least 5% less than its visible light transmission when irradiated from the uncoated side.
7. A multiple glazing unit as claimed in claim 5 wherein the coated outer pane has a total solar heat transmission at least 10% less than its visible light transmission.
8. A multiple glazing unit as claimed in any of the preceding claims wherein the coated outer pane has a visible light transmission of at least 60%.

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INTERNATIONAL SEARCH REPORT

International Application No

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A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C03C3/087 C03C4/02 C03C17/23

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C03C E06B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 616 883 A (SAINT GOBAIN VITRAGE) 28 September 1994	1-4, 6-9, 12-20, 22, 24-26
Y	see column 5, line 18 - line 30 see column 7, line 40 - line 52 see claims 1-7; figures 1-3 ---	5, 10, 11, 21, 23
X	EP 0 353 140 A (SAINT GOBAIN VITRAGE) 31 January 1990	1-4, 6-9, 12-20, 22, 24-26
Y	see column 4, line 57 - column 5, line 6 see column 5, line 61 - column 6, line 30 see column 7, line 3 - line 10 see claims 1-7; figures 1-3 ---	5, 10, 11, 21, 23
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

Int .tional Application No

PCT/GB 98/00407

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 192 009 A (SAINT GOBAIN VITRAGE) 27 August 1986 see example 1	1-26
A	& US 4 859 499 A (BONNAUD MICHELINE ET AL) 22 August 1989 see column 3, line 65 - column 4, line 24 see column 4, line 48 - line 51 see column 6, line 17 - line 27	1-26
X	FR 2 721 599 A (SAINT GOBAIN VITRAGE) 29 December 1995	1-8, 12-14, 16-26
Y	see page 2, line 24 - page 3, line 17 see page 7, line 1 - page 8, line 2 see page 8, line 15 - line 18 see page 8, line 24 - line 26 see examples 3,14	9-11,15
X	GB 2 068 442 A (BFG GLASSGROUP) 12 August 1981 cited in the application	1,2,5-8, 10,12, 13,16-26
Y	see page 2, line 28 - line 29 see page 3, line 6 - line 15 see page 5, line 6 - line 11; tables 1-7	3,4,9, 11,14,15
Y	GB 2 031 756 A (GORDON ROY GERALD) 30 April 1980 cited in the application	11
A	see the whole document	1-10, 12-26
X	EP 0 452 207 A (SAINT GOBAIN VITRAGE) 16 October 1991	1,2,5-9, 12,13, 15, 20-22, 24-26
Y	see page 2, line 58 - page 3, line 15 see page 4, line 18 - line 24; claims 1-12; table see page 3, line 57 - page 4, line 2	3,4,10, 11,14
A		16-19,23
A	EP 0 121 459 A (SAINT GOBAIN VITRAGE) 10 October 1984 see the whole document	
E	EP 0 825 478 A (SAINT GOBAIN VITRAGE) 25 February 1998 see claims	1,12
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INTERNATIONAL SEARCH REPORT

In tional Application No
PCT/GB 98/00407

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>EP 0 573 325 A (SAINT GOBAIN VITRAGE) 8 December 1993 see column 1, line 28 - line 33 see column 3, line 31 - column 4, line 10 -----</p>	1-26

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB 98/00407

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